Nature of c-axis coupling in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ with varying degrees of disorder

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The dependence of the Josephson Plasma Resonance (JPR) frequency in heavily underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ on temperature and controlled pointlike disorder, introduced by high-energy electron irradiation, is cross-correlated and compared to the behavior of the ab–plane penetration depth. It is found that the zero temperature plasma frequency, representative of the superfluid component of the c-axis spectral weight, decreases proportionally with T_c when the disorder is increased. The temperature dependence of the JPR frequency is the same for all disorder levels, including pristine crystals. The reduction of the c-axis superfluid density as function of disorder is accounted for by pair-breaking induced by impurity scattering in the CuO_2 planes, rather than by quantum fluctuations of the superconducting phase. The reduction of the c-axis superfluid density as function of temperature follows a T^2–law and is accounted for by quasi-particle hopping through impurity induced interlayer states.

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I. INTRODUCTION

Significant controversy remains concerning an appropriate model description of high temperature superconducting cuprates (HTSC) in the underdoped regime, *i.e.* the regime in which the number of additional holes per Cu, p is smaller than the value 0.16 at which the critical temperature T_c is maximum.[1] Whereas it is well established that the charge dynamics and transport properties in the normal- and superconducting states in the overdoped regime ($p > 0.16$) are, by and large, determined by well-defined quasi-particles, the role of quasiparticles in the underdoped regime is debated. The underdoped region of the (p, T) phase diagram is characterized by several salient features.[2] At $T^* > T_c$, the well-known "pseudo-gap" in the excitation spectrum opens up. This has been interpreted as either being related to the advent of another type of (spin- or charge-) order competing with superconductivity, driving T_c down as p is diminished, or, alternatively, as signalling the formation of precursor Cooper pairs without long-range phase coherence. Then, T_c is interpreted as the demise of long range superconducting phase order due to strong thermal[3,4,5] or quantum[6,7] phase fluctuations. Strong support for this scenario has come from the violation of the Glover-Tinkham-Ferrell conductivity sum-rule applied to the c-axis spectral weight;[5] also, the linear relation between T_c and the superfluid density[8] has been interpreted as the result of T_c being determined by phase fluctuations in a Kosterlitz-Thouless-type scenario.[9,10] A smoking gun for such a scenario would be an important reduction of the c-axis superfluid density ρ_s^c, which is determined by Cooper pair and quasiparticle tunneling between adjacent strongly superonducting CuO_2 layers through the weakly superconducting rocksalt-like blocking layers, with respect to the in-plane stiffness ρ_s^{ab} in underdoped cuprates *below* T_c.

However, apart from phase fluctuations, other mechanisms for the reduction of ρ_s^c, arising from disorder in the crystalline structure of underdoped cuprates, cannot be ignored.[11] First, scanning tunneling spectroscopy (STS) experiments[12,13,14] have revealed large variations of the magnitude of the gap maximum Δ_{peak}, as interpreted from conductance curves measured on the surface of $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals. This has motivated recent interpretations of weakened c-axis superfluid response in this material[15,16] as well as in $La_{2-x}Sr_xCuO_4$[17] in terms of finely dispersed $5 - 20$ nm-sized non-superconducting

regions within the CuO_2 planes. Such regions may arise from the suppression of the superconducting order parameter by dopant atoms,[18] such as out-of-plane oxygen atoms in the $Bi_2Sr_2CaCu_2O_{8+\delta}$ compound.[19]

Moreover, the d-wave symmetry of the gap function is at the basis of several mechanisms by which pointlike disorder reduces the c-axis superfluid density. The appearance of quasiparticle (virtual-) bound states and their smearing by a finite defect density leads to an increase of the density of states (DOS) near the nodal (π,π) directions (the so-called lifetime effect).[20] The same pointlike defects increase the quasiparticle scattering rate Γ_s. It was conjectured that in the case of coherent (in-plane momentum preserving) quasiparticle tunneling, the cancellation of these effects leads to disorder-independent low–temperature c-axis quasiparticle conductivity and superfluid density.[21] The approach of Ref. [21] neglects the crystal structure of the tetragonal HTSC,[11] which leads to the dependence of the interlayer hopping integral $t_\perp$ on the in-plane momentum (k_x, k_y). For simple tetragonal structures, interlayer hopping occurs via Cu 4s orbitals in adjacent planes. Its momentum dependence $t_\perp = t_\perp^0[\cos k_x a - \cos k_y a]^2$ is determined by the in–plane overlap of the bonding oxygen 2p level with the 4s level of the neighboring Cu atom.[22,23] As a result, c-axis tunneling occurs nearly exclusively for the anti-nodal directions at which quasiparticles are unlikely to be excited. In body-centered tetragonal structures such as $Bi_2Sr_2CaCu_2O_{8+\delta}$, hopping is also suppressed along the $(k_x, k_y) = (\pi, 0)$ and $(0, \pi)$ lines, yielding $t_\perp = t_\perp^0[\cos k_x a - \cos k_y a]^2 \cos\frac{1}{2}k_x a \cos\frac{1}{2}k_y a$.[24] In either case, disorder is always relevant for the nodal directions. Then, from the lifetime effect, one expects a quadratic decrease with temperature of the reduced c-axis superfluid density[23,25]

$$\frac{\rho_s^c(T)}{\rho_s^c(0)} \propto 1 - \alpha_c \frac{8\pi}{3}\frac{\Gamma_s}{\Delta_0}\left(\frac{T}{\Delta_0}\right)^2, \qquad (k_B T \ll \Gamma_s). \quad (1)$$

Here α_c is a dimensionless constant of order unity and the parameter Δ_0 was assumed, in Refs. 23 and 25, to correspond to the maximum amplitude of the Bardeen-Cooper-Schrieffer d-wave gap. Eq. (1) essentially differs from that derived for the ab-plane superfluid density

$$\frac{\rho_s^{ab}(T)}{\rho_s^{ab}(0)} \propto 1 - \alpha_{ab} \frac{\Delta_0}{\Gamma_s}\left(\frac{T}{\Delta_0}\right)^2, \qquad (k_B T \ll \Gamma_s) \quad (2)$$

in that the leading temperature–dependent term has a coefficient that is smaller by a factor $(\Gamma_s/\Delta_0)^2$.[23] The presence of defects in the rocksalt-like (BiO) layers tends to break the d-wave symmetry of the hopping integral, and renders quasiparticle hopping possible for other values of the in-plane momentum, and notably along the order parameter nodes.[11,23,25,26] A condition for this "impurity-assisted hopping" (IAH) to be effective is an anisotropic scattering matrix of the interplane defects. Notably, for strong forward scattering, the result

$$\rho_s^c(T) \approx 2\pi V_1 \Delta_0 N^2(E_F)\left[1 - 8\ln 2\left(\frac{T}{\Delta_0}\right)^2\right], \quad (3)$$

was obtained for $\Gamma_s \ll k_B T \ll \frac{1}{2}\left[2\pi V_1 \Delta_0 N(E_F)/(t_\perp^0)^2\right]^{1/3} T_c$.[23,27] Here V_1 is the magnitude of the impurity scattering potential of the out-of-plane defects and $N(E_F)$ is the density of states at the Fermi level in the normal state. The effect of impurities can be distinguished from that of boson-assisted interlayer hopping; for the latter, a very similar result is obtained, but with the leading temperature–dependent term proportional to T^3.[25,28] Finally, direct hopping of quasiparticles was suggested to lead to a small, linearly temperature-dependent, reduction of ρ_s^c.[27]

In this paper, we address the mechanism by which the c-axis superfluid density in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (with $p = 0.10$) is reduced by using disorder, in the form of Frenkel pairs introduced by high energy electron irradiation, as an independent control parameter. Electron irradiation, the effects of which are taken to be similar to those of Zn-doping,[29] has previously been used to study the effect of pointlike disorder on the resistivity, critical temperature,[30] and Nernst effect of $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.6}$.[31] In the latter material, electron irradiation eventually leads (at high fluences) to the breakdown of the well-known Abrikosov Gor'kov relation[32,33]

$$\ln\left(\frac{T_c}{T_{c0}}\right) = \Psi\left(\frac{1}{2}\right) - \Psi\left(\frac{1}{2} + \frac{\Gamma}{2\pi k_B T_c}\right) \quad (4)$$

(with T_{c0} the critical temperature when the normal state scattering rate Γ is equal to zero, and Ψ the digamma function) as well as a significant increase of the fluctuation regime near T_c.[31] Both effects were interpreted as the effect of strong superconducting phase fluctuations.[30,31] The in-plane and c-axis superfluid densities of Zn-doped $YBa_2Cu_3O_{7-\delta}$ were studied by Panagopoulos *et al.*[34] and by Fukuzumi, Mizuhashi, and Uchida.[35] The progressive inclusion of Zn leads to a rapid decrease of the in-plane superfluid density ρ_s^{ab}, corresponding to an increase of the in-plane penetration depth $\lambda_{ab}(0) \propto (\rho_s^{ab})^{-1/2}$, and a more modest decrease of $\rho_s^c \propto \sigma_c(T_c) \propto T_c$, that violates the c-axis conductivity sum-rule[5,35] [$\sigma_c(T_c)$ is the c-axis conductivity at T_c]. As for the low–T temperature dependence, a gradual change of both ρ_s^{ab} and ρ_s^c from T–linear to T–squared has been reported.[34] Studies on $Bi_2Sr_2CaCu_2O_{8+\delta}$ are limited to electron irradiation of the single crystalline optimally doped material, that show a linear decrease of T_c with electron fluence.[36,37,38] The c-axis superfluid density in a underdoped pristine $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal has been previously studied by Gaifullin *et al.*, who invoked the IAH model to explain the much stronger temperature dependence of ρ_s^c in underdoped with respect to optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$.[39]

Below, we report on c-axis coupling in the superconducting state measured through the Josephson Plasma Resonance (JPR),[40,41,42,43] which, in our underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals takes place in the microwave frequency regime below 70 GHz. The JPR frequency f_{pl} is sensitive to the value of Δ_0, as well as to fluctuations of the superconducting order parameter phase in the CuO_2 planes.[6] The evolution of $f_{pl}(T)$ with temperature depends simultaneously on the quasiparticle dynamics and on the strength of fluctuations; the plasma resonance peak is broadened both by the quasiparticle tunneling rate and by crystalline disorder.[17,44] However, the dependence of $f_{pl}^2 \propto \rho_s^c$ on the disorder strength is expected to be quite different, depending on which mechanism is predominant. In the following, we show that the disorder dependence of the c-axis plasma frequency is a sensitive probe, that allows one to identify in detail what physical mechanisms are at the basis of the reduction of the superfluid density in $Bi_2Sr_2CaCu_2O_{8+\delta}$. It turns out that, even in our heavily underdoped crystals, (incoherent) c-axis quasiparticle hopping is essential for a consistent description of the data. We find that the energy scale Δ_0, which turns out to be $Delta_0 \approx 2.5 k_B T_c$ for all underdoped crystals, is to be interpreted as an energy scale governing nodal quasi-particle excitations.

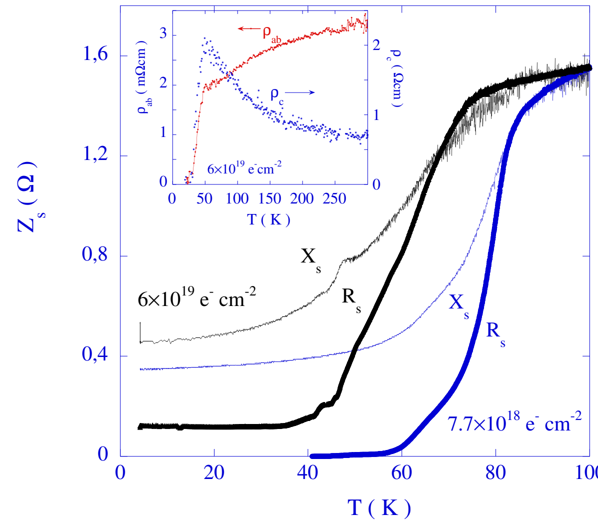

FIG. 2: (color online) Real (R_s) and imaginary (X_s) parts of the surface impedance Z_s of underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals, irradiated with 7.7×10^{18} and 6×10^{19} electrons cm^{-2}, respectively. The data were obtained from the resonance frequency shift and the quality factor of a superconducting Pb cavity operated in the TE_{011} mode. The inset shows the ab–plane– and the c-axis dc resistivity of the crystal irradiated with 6×10^{19} electrons cm^{-2}.

II. EXPERIMENTAL DETAILS

The underdoped ($T_c = 65 \pm 0.5$ K, $p \approx 0.10$) $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals, of typical dimensions $500 \times 300 \times 40 \mu m^3$, were selected from the same boule, grown by the travelling solvent floating zone method at the FOM-ALMOS center, the Netherlands, in 25 mBar O_2 partial pressure.[45] The crystals were annealed for one week in flowing N_2 gas. We have also measured a set of optimally doped control samples ($T_c = 86$ K). These were also grown by the travelling solvent floating zone technique, at 200 mbar oxygen partial pressure, and subsequently annealed in air at 800°C. The crystals were irradiated with 2.3 MeV electrons using the Van de Graaff accelerator at the Laboratoire des Solides Irradiés. The beam was directed along the crystalline c-axis during the irradiation. To prevent recombination and clustering of point defects, the irradiation is carried out with the crystals immersed in a liquid hydrogen bath (22 K). The electron flux is limited to 2×10^{14} e$^-$cm^{-2} per second. Crystals UD5-UD8 were irradiated to a total fluence of 0.53×10^{18}, 3×10^{18}, 7.7×10^{18}, and 8.8×10^{19} e$^-$cm^{-2} respectively. After measurements, crystal UD5 was irradiated a second time to a total fluence of 6.0×10^{19} e$^-$cm^{-2} and was henceforth labeled UD5b. The high energy electron irradiation creates random atomic displacements in the form of Frenkel pairs, both in the CuO_2 bilayers and in the intermediate cation layers, throughout the samples.

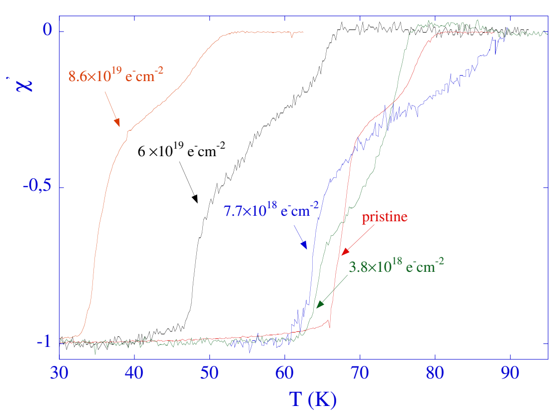

FIG. 1: (color online) Real part of the ac magnetic susceptibility of the investigated underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals. The crystals were irradiated at 22 K with 2.3 MeV electrons to the indicated fluences. The ac field amplitude $h_{ac} = 4.2$ mOe, the ac frequency was 560 Hz. The curves show a screening onset determined by a surface layer containing optimally doped material; this screening is suppressed when $h_{ac} \gtrsim 0.5$ Oe. The steep drop at the lower end of the transitions corresponds to bulk screening by the underdoped material.

The superconducting transition temperature T_c was determined by ac susceptibility measurements using a driving field of amplitude 4.2 mOe and a frequency of 560 Hz, directed parallel to the c-axis. For all underdoped crystals, the superconducting transition is rather

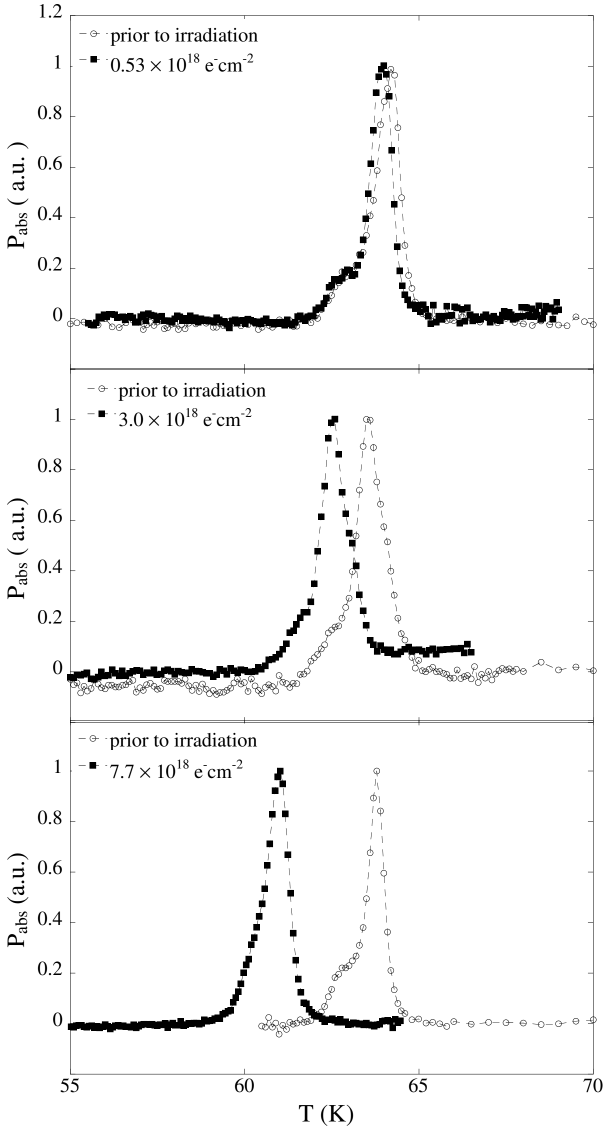

FIG. 3: Microwave absorption, measured using the TM_{010} mode (19.2 GHz) of one of the OFHC copper cavities, of three of the underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals before and after irradiation with 5.3×10^{17}, 3×10^{18}, and 7.7×10^{18} 2.3 MeV electrons cm^{-2}, respectively.

broad. The superconducting transition takes place in two steps: there is a slow increase of screening at high temperature, followed by a rapid step of the diamagnetic signal at lower temperature (Figure 1). The high temperature screening vanishes when the excitation field amplitude is increased beyond 0.5 Oe, while the step at lower temperature is robust. This shows that doping is macroscopically inhomogeneous, and that the crystals are surrounded by a thin surface layer of higher doping. This layer could not be eliminated by cutting the crystals. The overall shape of the transition is unaffected by the electron irradiation. The transition widths are of the order of 4 K, which is usual for such low doping. After irradiation, the transition widths slightly increase. For all crystals, the transition to zero dc resistivity and bulk superconductivity occurs at the temperature at which the lower screening step takes place, see *e.g.* the Inset to Fig. 2. Therefore, the lower temperature feature was adopted as characterizing the bulk T_c of the underdoped crystals.

Crystals were further characterized by the measurement of the temperature variation of the in-plane penetration depth, $\lambda_{ab}(T)/\lambda_{ab}(0) - 1 \equiv \Delta\lambda_{ab}/\lambda_{ab}$. For this, a crystal is mounted on a sapphire rod, in the center of a superconducting (Pb) resonant cavity immersed in liquid ^{4}He, and operated in the TE_{011} mode. The cavity resonant frequency was $f \sim 27.8$ GHz, and the quality factor $Q \sim 4 \times 10^5$. The crystal is mounted in such a way that the magnetic microwave field is perpendicular to its ab plane and solely in-plane screening currents are induced. From the shift Δf of the cavity resonance frequency induced by the sample, we determine the surface reactance $X_s = 2\pi\mu_0 G_2 \Delta f$ (with $\mu_0 = 4\pi \times 10^{-7}$ Hm^{-1}). The surface resistance $R_s = 2\pi\mu_0 G_1 (\Delta Q)^{-1}$ was obtained from the change of the quality factor. The geometrical factors G_1 and G_2 were determined by comparing the surface impedance in the normal state, $X_s = R_s = \pi\mu_0 f \delta_s$, to the value expected from the normal state resistivity, $\rho = \pi\mu_0 f \delta_s^2$.[46] It was retrospectively checked that all measurements were carried out in the skin effect regime, in which the normal state skin depth δ_s is much smaller than the sample dimensions. The relative change of the penetration depth was determined from the behavior of the surface reactance at low temperature, at which $X_s \approx 2\pi\mu_0 f \lambda_{ab}$.

Figure 2 shows that the temperature at which the decrease of the surface resistance R_s was observed corresponds to the (high temperature) onset of screening in the ac susceptibility measurement. This indicates that the surface skin depth of thickness $\sim 7\mu$m probed by the microwave field contains patches with larger hole content p.

The JPR measurements were performed using the cavity perturbation technique, using the TM_{01n} modes ($n = 0, ..., 4$) of two Oxygen-Free High Conductivity (OFHC) copper resonant cavities ($Q \sim 4000 - 10000$), mounted on a cryocooler cold head. The measurement frequencies ranged between 19.2 and 39.6 GHz.[47] Further measurements were made applying the bolometric technique, using waveguides in the TE_{01} travelling wave mode.[39] In both measurement set-ups, the electrical microwave field is applied along the c-axis of the crystal. In contrast to the previously described surface impedance measurements, screening of the electric microwave field is very poor because of the high electronic anisotropy of the crystals. The underdoped samples are in the complete depolarization regime and thus the bulk electromagnetic response is probed. By monitoring the power absorption as a function or frequency for a fixed temperature, the Josephson Plasma Resonance is detected as a sharp absorption peak in the microwave response, see Fig. 3. We determine the JPR frequency at a given temperature,

$f_{pl}(T)$, as the measurement frequency at the temperature at which dissipation is maximum.

III. RESULTS

Figure 4 collects the values of the critical temperature as function of electron dose, for the set of underdoped samples as well as the optimally doped control samples. Both the T_c of the underdoped and the optimally doped crystals decrease linearly with irradiation fluence. The derivative of T_c with respect to fluence of the optimally doped crystals concurs with that measured by Behnia *et al.*[37] and Nakamae *et al.*[38] but is two times lower than that measured by Rullier-Albenque *et al.*.[36] The overlap between the variation with fluence of the screening onset temperature in the underdoped crystals and the T_c of the optimally doped crystals shows that the thin surface layer on the underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ has $p \sim 0.16$. The critical temperatures, normalized to the critical temperatures T_{c0} of the unirradiated crystals, can be superimposed on Eq. (4), yielding estimates of the normal state scattering rate Γ (see Fig. 5). This procedure supposes that T_{c0} corresponds to the critical temperature in the absence of disorder; we shall see below that this is not justified, so that the estimated Γ values are in fact lower limits for each crystal.

The relative change with temperature of the in-plane penetration depth λ_{ab} is depicted in Fig. 6. For all underdoped crystals, including the unirradiated ones,

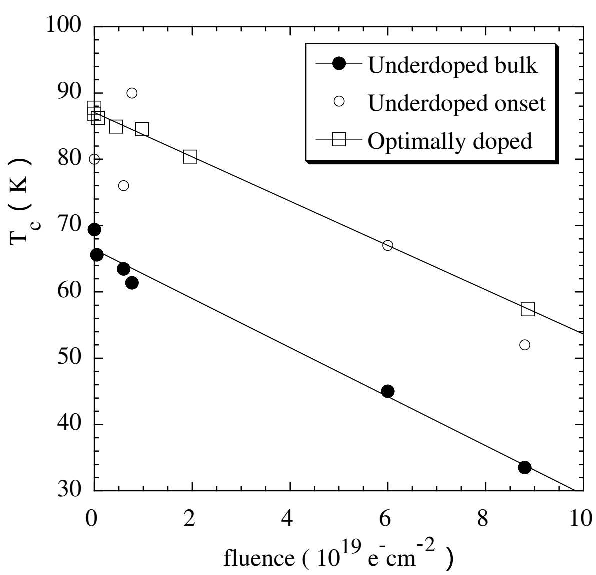

FIG. 4: Variation of the critical temperature with electron fluence for single-crystalline underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ ($\bullet$). A comparison with the T_c of a series of optimally doped control crystals ($\square$) shows that the onset temperature of magnetic screening ($\circ$) corresponds to flux exclusion by a surface region containing optimally doped material.

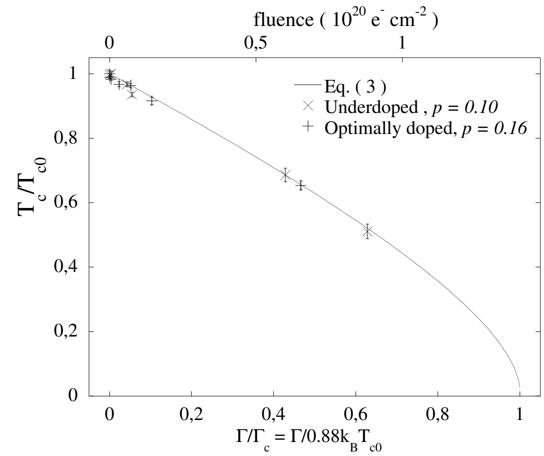

FIG. 5: Critical temperatures T_c normalized to the critical temperature T_{c0} of the unirradiated crystals, and superimposed on the Abrikosov-Gor'kov relation (4). Note that the actual data points should be shifted downwards along the curve, because T_{c0} does not truly correspond to the critical temperature in the absence of disorder.

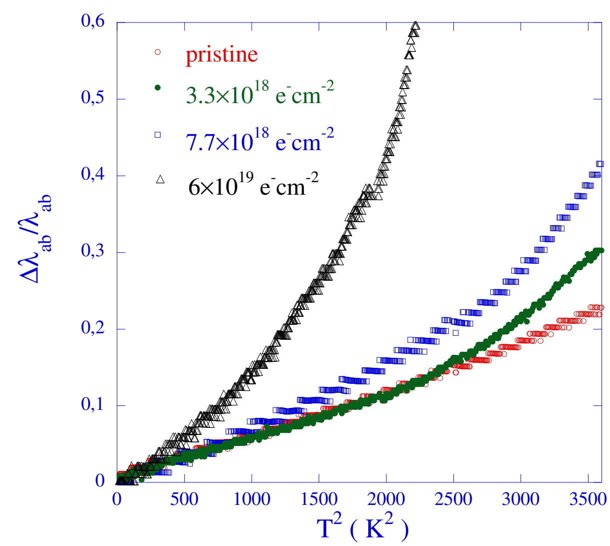

FIG. 6: (color online) Relative change of the ab–plane penetration depth $\Delta\lambda_{ab}/\lambda_{ab}(0) = [X_s(T)/X_s(T \rightarrow 0) - 1]/2\pi\mu_0 f$, as obtained from the change in the inductive part X_s of the surface impedance.

$\lambda_{ab}(T) - \lambda_{ab}(0)$ varies quadratically with temperature at low T. Such a temperature dependence has been associated with quasiparticle scattering in the unitary limit by point defects situated within the CuO_2 planes of the d-wave superconductor, *i.e.* $N(E_F)V \gg 1$, $\Gamma \sim n_d/\pi N(E_F)$, and $\Gamma_s \sim 0.6(\Gamma\Delta_0)^{1/2}$.[48] Here V is the scattering potential of the defects *in* the planes, with density n_d. The quadratic temperature dependence of $\Delta\lambda_{ab}(T)$ is at odds with a possible important role of thermal phase fluctuations, for which a linear T–dependence was

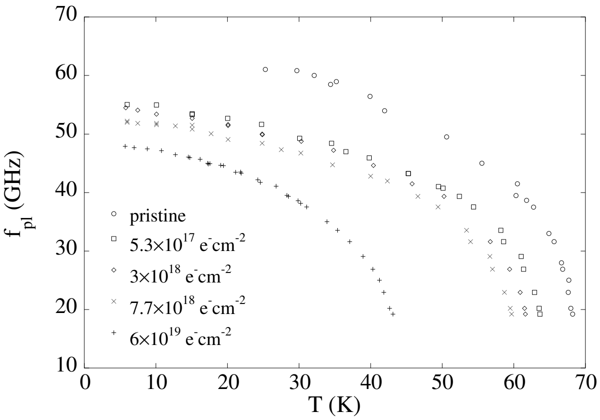

FIG. 7: Temperature dependence of the JPR frequency of electron-irradiated underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$.

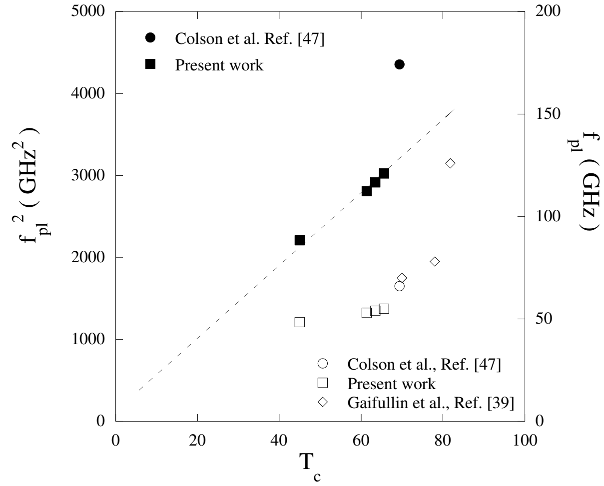

FIG. 8: $f_{pl}(0)$ versus T_c (open symbols), as well as $f_{pl}^2(0) \propto \rho_s^c(0) \propto j_c^c(0)$ versus T_c (closed symbols), for both the underdoped irradiated crystals (squares) and a set of crystals with different doping levels ($p \approx 0.13$,[39] and $p \approx 0.11$[47]).

predicted.[49] As for the magnitude of the T^2-contribution to λ_{ab}, a very modest change is found for the lower irradiation fluences. Only for fluences exceeding 10^{19} e$^-$cm^{-2} does the in-plane penetration depth increase significantly with defect density.

We now switch to the central results concerning the Josephson Plasma Resonance. Figure 7 shows the JPR frequency $f_{pl}(T)$ of crystals UD5-UD7 as function of temperature (measured in Earth's magnetic field). The temperature at which $f_{pl}(T)$ extrapolates to zero is well-defined and corresponds to the critical temperature of the bulk, underdoped portion of the crystals, *i.e.* the main transition in the ac susceptibility and zero resistance. This shows that the JPR probes the c-axis response in the heavily underdoped bulk and is insensitive to the surface quality of the samples. From Fig. 7 one sees that not only T_c, but also f_{pl} is strongly depressed by the electron irradiation. Fig. 8 collects values of the low-temperature extrapolated value $f_{pl}(0)$ versus the critical temperature, and reveals the proportionality between $f_{pl}^2(0)$ and T_c. This dependence is clearly different from the variation of f_{pl} with oxygen doping. The same Figure recapitulates results for doping levels $p = 0.13$[39] and 0.11,[47] the evolution of which recalls the exponential $f_{pl}^2(0)(T_c)$–dependence found by Shibauchi *et al.*.[16] The results are somewhat similar to those obtained by Fukuzumi *et al.* for Zn-doped and oxygen-deficient $YBa_2Cu_3O_{7-\delta}$:[35] however, their Fig. 5 shows a linear dependence of f_{pl} on T_c for underdoped $YBa_2(Cu_{1-x}Zn_x)_3O_{6.63}$, and a hyperbolic or exponential evolution of $f_{pl}(T_c)$ as function of δ.

As for the temperature variation of $f_{pl}^2(T) \propto \rho_s^c$, it turns out to be identical for all the underdoped crystals, including the pristine ones, and does not depend on the defect density. Fig. (9) shows $f_{pl}^2(T)/f_{pl}^2(0)$ plotted versus the reduced temperature T/T_c. For $hf_{pl} < \Gamma$, this is representative of the c-axis superfluid fraction ρ_s^c. The same graph may also be interpreted as the maximum Josephson (c-axis) critical current,

$$j_c^c = 2\pi(h/2e)\epsilon\epsilon_0 s^{-1} f_{pl}^2 = \frac{h}{4\pi e\mu_0\lambda_c^2 s} \qquad (5)$$

normalized to its value for $T \to 0$. Here s is the spacing between CuO_2 planes, the c-axis dielectric constant $\epsilon \approx 11.5$,[50] and $\epsilon_0 = 8.854 \times 10^{-12}$ Fm^{-1}.

IV. DISCUSSION

To distinguish between the different mechanisms responsible for the reduction of the c-axis plasma frequency as temperature is increased, we dispose of three tools. First, there is the variation of $f_{pl}^2(0)$ with disorder strength, which manifests itself starting from the smallest electron fluences, and proportionally follows the evolution of T_c with disorder. Second is the temperature variation $f_{pl}^2(T)/f_{pl}^2(0)$, that follows a $1 - a(T/T_c)^\alpha$ dependence, with $\alpha \sim 2$ independent of the disorder strength. Finally, there is the comparison with the behavior of the in-plane penetration depth, $\lambda_{ab}(T)/\lambda_{ab}(0) \sim 1 + \beta T^2$. A successful model description should account for all three dependences correctly.

The theoretically expected low-temperature dependence $\lambda_c(T)$ depends strongly on a number of circumstances. First is the question whether superconductive coupling is three-dimensional (*i.e.* the c-axis momentum k_z is a good quantum number),[23] or whether it is mediated by Josephson tunneling between CuO_2 layers. Josephson coupling can be weakened by direct[21,27] or boson-assisted quasiparticle tunneling,[25,27,28] or by tunneling that involves intermediate defect-induced states between the layers (IAH).[23,27] For "direct" tunneling,

e.g. occurring through direct overlap of the superconducting wave functions in the CuO_2 planes, one distinguishes the case of conserved in-plane momentum $k_\parallel$ ("coherent" tunneling) from the case where it is not preserved ("incoherent" tunneling[21] — this situation would yield a vanishing j_c^c in a d–wave superconductor). A prevailing effect of nodal quasiparticles leads to a more rapid decrease of j_c^c , see Eq. (1). Finally, in all cases the anisotropy of the transfer matrix $t_\perp$ is expected to have an important influence on the temperature dependence of λ_c,[23,52] notably reconciling a weak T–dependence of λ_c with a strong variation of $\lambda_{ab}(T)$. Here, the experimentally observed temperature dependence of $f_{pl}^2(T)/f_{pl}^2(0)$ actually allows one to discard a dominant role of a possible d-wave symmetry of the transfer matrix $t_\perp$,[23] since (for $k_BT \gg \hbar\Gamma_s$) this leads to a weak temperature dependence, $f_{pl}^2(T)/f_{pl}^2(0) \sim 1 - \tilde{a}T^5$, observed in slightly overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$[39,50] and optimally doped $HgBa_2Ce_{n-1}Cu_nO_{2n+2+\delta}$,[51] but not in the present data on underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$. The modification of $t_\perp$ arising from the body-centered Bravais lattice of the $Bi_2Sr_2CaCu_2O_{8+\delta}$ compound will influence the maximum c-axis critical current. However, it will not change the expected $1-\tilde{a}T^5$ temperature dependence, since this arises from the specific coincidence in k-space of the zero of $t_\perp(k_x,k_y)$ with the nodal direction of the order parameter. Thus, models for superconductive coupling,[23] or direct Josephson coupling with a vanishing hopping integral along the nodal line[52] are in inadequacy with the data.

We next exclude a dominant role of direct quasiparticle tunneling. Even though the similar T^2–dependences of the low–temperature ab–plane and c-axis penetration depths suggests such coupling, the disorder dependence is at odds with the experimental result. Radtke *et al.*[27] and Latyshev *et al.*[21] find that for direct coupling, the low temperature c-axis critical current

$$j_c^{c,direct} = \frac{\pi\sigma_q^c(0)\Delta_0}{es} = \frac{4\pi e t_\perp^2 N_n(E_F)}{h} \qquad (6)$$

is, for $k_BT \ll \hbar\Gamma_s \sim 20-30$ K, to lowest order independent of the defect density due to the cancellation of the scattering-rate dependences of the quasi-particle conductivity σ_q^c and Δ_0. The model was further worked out by Kim and Carbotte, who find that to first order

$$j_c^{c,direct} \propto 1 - \alpha\frac{\Gamma_s}{\sqrt{\Gamma_s^2+\Delta_0^2}} \sim 1 - \alpha\frac{1}{\sqrt{1+\Delta_0/\Gamma}} \qquad (7)$$

both for the case of constant $t_\perp$ (where $\alpha \approx 1$) and angular–dependent $t_\perp$ ($\alpha = \frac{16}{9}$).[11] The nonlinear dependence (7) is at odds with the observed linear evolution of j_c^c with irradiation fluence.

The temperature and disorder-dependence of the low temperature c-axis JPR frequency is more successfully described by a model for incoherent tunneling. According to Latyshev *et al.*, an incoherent tunneling process yields $j_c^{c,i} \approx j_c^{c,direct}\Delta_0/E_F \approx 4\pi e t_\perp^2 N_n(E_F)\Delta_0/hE_F$. The extra factor Δ_0 then explains the linear relation between

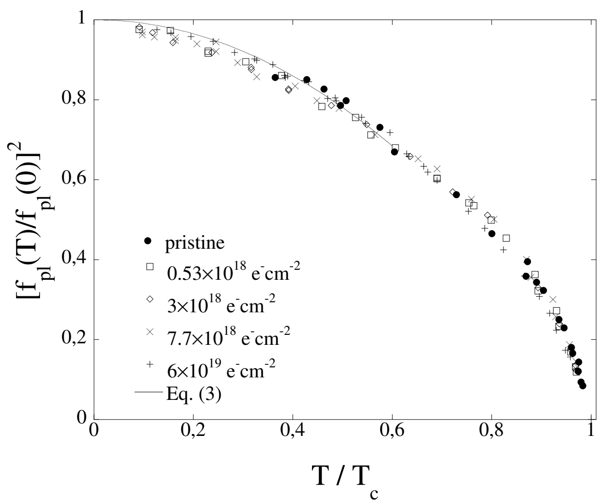

FIG. 9: Square of the JPR frequency, normalized to its low temperature extrapolation $f_{pl}(0)$, versus reduced temperature T/T_c. This plot is representative of the temperature dependence of the c-axis superfluid stiffness, as well as of the maximum c-axis Josephson current: $f_{pl}^2(T)/f_{pl}^2(0) \sim \lambda_c^2(0)/\lambda_c^2(T) \sim \rho_s^c(T)/\rho_s^c(0) \sim j_c^c(T)/j_c^c(0)$. The drawn line is a fit to Eq. (3) with $\Delta_0 = 2.5k_BT_c$.

$j_c^c(0)$ and T_c, accepting that in a d-wave superconductor with impurity scattering in the unitary limit, $\Delta_0(\Gamma)$ is simply proportional to $T_c(\Gamma)$.[33] The linear dependence on Δ_0 is found in the IAH model, see Eq. (3).[23,27,28] The latter expression consistently describes the fact that the temperature dependence of f_{pl} does not change with defect density: the temperature dependent term writes $(T/\Delta_0)^2 \propto (T/T_c)^2$. For the same reason, the "lifetime effect", Eq. (1), does not describe the data: in the unitary limit, the leading temperature–dependent term has an extra factor $\Gamma_s/\Delta_0 \sim (\Gamma/\Delta_0)^{1/2}$ and is therefore expected to strongly depend on defect density. The observed defect–density independence of $\rho_s^c(T)/\rho_s^c$ would require the strength of the scattering potential of the individual irradiation defects in the CuO_2 planes to be in the Born limit, which contradicts the results on the temperature dependence of the ab–plane penetration depth. We note that the toy model for incoherent hopping of Ref. [11], which yields $\lambda_c^{-2} \propto [1 - \frac{5}{14}(\Gamma_s/\Delta_0)^2 - \ldots] \sim [1 - \frac{3}{14}(\Gamma/\Delta_0) - \ldots]$, also describes the initial linear decrease of $j_c^c(T_c)$ (Eq. (31) of Ref. [11]).

Given the success of the IAH–model in qualitatively explaining the temperature- as well as the disorder dependence of the JPR data, we perform a direct fit of $f_{pl}^2(T)/f_{pl}^2(0)$ to Eq. (3), shown in Fig. 9. The only parameter is the value $\Delta_0 = 2.5k_BT_c$. This value not only means that $\Gamma_s > 30$ K, comforting our interpretation of the decrease of $j_c^c \propto T_c$ in terms of unitary scatterers induced in the CuO_2 planes, it is also remarkably close to characteristic energy scales found in recent Raman scattering[53] and STM experiments.[54] The first

study finds that in underdoped $HgBa_2CuO_4$, the B_{2g} Raman mode, which directly couples to the same low-energy nodal quasi-particle excitations that are responsible for the reduction of the c-axis superfluid density in the IAH model, is characterized by an energy scale $\sim 2k_BT_c$ (related to the nodal slope of the gap function).[53] We surmise that Δ_0 is a closely related parameter. The second study finds that the total tunneling gap amplitude is determined by two energy scales, the smaller of which, $\Delta_\phi \approx 2.8k_BT_c$, is related to superconductivity.[54] It is interesting that the existence of a second small energy scale describing nodal quasiparticle exitations may well be responsible for the observed suppression of j_c^c as function of doping (decreasing p or δ).[16] The proportionality of this decrease to the ratio of the gap maximum (at the antinode) and T_c finds a logical explanation if the smaller energy scale ($\sim 2 - 2.5k_BT_c$) is what determines the magnitude of the c-axis critical current density.

Thus, the (incoherent) interlayer assisted hopping is a feasible candidate for the reduction of the c-axis superfluid density in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$: it parametrically describes the data, and numerical values extracted for the relevant energy scale determining the quasiparticle excitations is the same as found in other experiments. The model does have several caveats: first, there is the above–mentioned puzzle that it requires the temperature dependence of the ab–plane superfluid density to be explained by the lifetime effect, whereas the same effect does not seem to play a role in the c-axis superfluid density, other than providing the quasiparticles. The second is the identification of the defects in the rocksalt–like layers that are responsible for interlayer scattering. The model by Xiang *et al.*[23,25] requires these out-of-plane defects to be weakly scattering, with a strongly anisotropic potential that leaves the reflected wavevector close to the incident ("strong forward scattering"). Although candidates may be out-of-plane oxygen defects[19] or cation disorder, the constraints imposed on the scattering potential seem very strict. In the end, the agreement of defect-density independence of the experimental $f_{pl}(T)/f_{pl}(0)$ with the IAH prediction[23] may be completely fortuitous. The coincidence of the temperature dependence of f_{pl} of the pristine crystals with even the most heavily irradiated ones indicates that disorder plays an important role in *all* samples. Notably, we expect T_c of pristine crystals to be substantially suppressed with respect to that of hypothetically "clean" underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$. Furthermore, impurity scattering is likely to completely suppress *any* role of quasi-particles in the c-axis electromagnetic response of this compound, leaving only pair tunneling.[6,55]

The remaining mechanism for the reduction of the superfluid density in the presence of pair tunneling only is that of order parameter phase fluctuations in the CuO_2–layers. The effect of quantum phase fluctuations on the ab–plane and c-axis superfluid densities was examined by Paramekanti *et al.*,[6,7] who performed an analytical study of an XY model for the superconducting order phase ϕ on a two-dimensional (2D) lattice of spacing ξ_0 (representing the coherence length), within the self-consistent harmonic approximation. The authors conclude that, in contrast to thermal phase fluctuations, quantum phase fluctuations in quasi 2D high temperature superconductors are important at all temperatures. The low carrier density in these materials leads to inefficient screening of the Coulomb interaction between charge carriers, and a sizeable reduction of the magnitude of the ab–plane superfluid density (without change of its temperature dependence). Within this model, the JPR frequency is also renormalized downwards because of fluctuations of the phase in the layers:

$$f_{pl}^2(T) = f_{pl}^2(0)\mathrm{e}^{-\langle \frac{1}{2}\phi_\perp^2 \rangle} \approx f_{pl}^2(0)\left(1 - \frac{1}{2}\langle \phi_\perp^2 \rangle - \ldots \right). \tag{8}$$

Paramekanti *et al.* estimate the phase difference between two points separated by a vector perpendicular to the superconducting layers as $\langle \phi_\perp^2 \rangle \approx \sqrt{e^2/4\pi\epsilon\epsilon_0\xi_0\varepsilon_0(0)s}$,[7] with $\varepsilon_0 s = h^2 s/16e^2\pi\mu_0\lambda_{ab}^2(0)$ the in–plane phase stiffness. We observe that, given the Uemura relation $\varepsilon_0 s \propto k_BT_c$,[8] the resulting expression naturally describes the experimentally observed exponential depression of j_c^c as function of doping.[16] However, even if one explicitly develops the dependence of $\varepsilon_0 s$ in terms of the variance of the *in-plane* phase $\langle \phi_\parallel^2 \rangle \sim \langle \phi_\perp^2 \rangle$, Eq. (8) fails to describe the linear $f_{pl}^2(T_c)$–dependence (Fig. 8). Therefore, the reduction of ρ_s^c with increasing disorder cannot be ascribed to quantum phase fluctuations only – pair-breaking in the CuO_2 layers must play a significant role.

A noteworthy prediction of the quantum fluctuation scenario is that the temperature evolution of the c-axis superfluid density is entirely determined by that of the in-plane phase stiffness, *i.e.* the c-axis and ab–plane superfluid densities follow the same dependence at low temperature. Inserting the experimental result $\lambda_{ab} = \lambda_{ab}(0)(1 + \beta T^2)$ into the prediction of Ref. [7], one would expect

$$\frac{\partial\left[f_{pl}(T)/f_{pl}(0)\right]^2}{\partial(T/T_c)^2} = -\frac{C_1}{4}\beta T_c^2\sqrt{\frac{2\pi e^2}{4\pi\epsilon\epsilon_0\xi_0\varepsilon_0(0)s}}, \tag{9}$$

This is experimentally verified; taking the data of Fig. 6 and $\lambda_{ab}(0) \approx 300$ nm,[47] we find that Eq. (9) is obeyed with $C_1 \approx 0.3$ (C_1 should be order unity[7]). The experimental independence of $f_{pl}(T)/f_{pl}(0)$ on defect density demands that $\beta T_c^2 \lambda_{ab}(0)$ is disorder independent. Adopting the generally accepted view that $\lambda_{ab}^{-2}(T)$ is described by Eq. (2) with Γ_s in the unitary limit, this would imply that $\beta T_c^2 \lambda_{ab}(0) \approx \alpha_{ab}\lambda_{ab}(0)(T_c/\Gamma)^{1/2}(T_c/\Delta_0)^{3/2}$, and therefore that $\lambda_{ab}(0)^{-2} \propto T_c/\Gamma$. This is as yet unverified, as the different sizes and aspect ratios of our crystals prohibit a direct comparison of the absolute values of λ_{ab}.

Note that the case of screening by nodal quasiparticles

was also studied in Ref. [7]. Then,

$$\frac{\partial\left[f_{pl}(T)/f_{pl}(0)\right]^2}{\partial(T/T_c)^2} \approx -\frac{2\beta T_c^2}{\overline{\sigma}_q}, \tag{10}$$

where $\overline{\sigma}_q$ is the ab–plane quasiparticle sheet conductivity, normalized to the quantum conductivity e^2/h. This formula also describes the temperature dependence of the data, provided that $\overline{\sigma}_q \approx 3$; moreover, the ratio $\beta/\overline{\sigma}_q \sim \alpha_{ab}\Delta_0(T_c/\Delta_0)m/N_s(0)e^2$ should be disorder–independent ($N_s(0)$ is the quasiparticle density of states and m the effective mass). Given the theoretical expectation $N_s(0) \sim \Gamma^{1/2}$ [48] and $\Delta_0 \propto 1-\Gamma$ [33], this model again fails to describe the reduction of the zero temperature c-axis superfluid density in terms of quantum fluctuations only.

V. SUMMARY AND CONCLUSIONS

We have cross-correlated the dependence of the c-axis Josephson Plasma Resonance frequency in heavily underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ on temperature and controlled disorder (introduced by high energy electron irradiation), and compared both with the behavior of the in-plane penetration depth. It is found that the c-axis critical current is depressed with increasing disorder strength, proportionally to the critical temperature T_c. Both the in–plane– and out–of–plane superfluid densities follow a T^2 temperature dependence at low T. The temperature dependence of the c-axis response is independent of disorder, indicating that we are probing the superfluid density. The superfluid response of the pristine underdoped crystals is indistinguishable from that of heavily irradiated ones, suggesting that pristine underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ commonly contains sufficient disorder in the CuO_2 planes for the critical temperature to be significantly suppressed with respect to what the T_c of the hypothetically "clean" material would be. The dominating in-plane disorder in as-grown crystals is likely to be of the same kind as introduced by the electron irradiation. Apart from unitary scatterers in the CuO_2 planes, this also encompasses the "order parameter holes" induced by dopant oxygen and cation disorder in the rocksalt-like layers.[18,19]

The experimental data were confronted with a variety of theoretical models describing the reduction of c-axis superfluid density in terms of either quasiparticle dynamics or quantum fluctuations of the superconducting order parameter phase in the CuO_2 layers. We find that the quantum phase fluctuation description[6,7] yields excellent agreement as to the experimentally observed similar temperature dependences of λ_{ab} and λ_c, and quantitatively describes the temperature derivative $\partial[f_{pl}(T)/f_{pl}(0)]^2/\partial(T/T_c)^2$. However, it fails to describe the dependence of the zero-temperature Josephson Plasma frequency on disorder strength.

We therefore surmise that the reduction of $f_{pl}(0)$ with increasing disorder must be due to pair-breaking within the CuO_2 layers. Data for $\lambda_{ab}(T)$ and $\lambda_c(T,\Gamma)$ are in agreement with scattering in the unitary limit by the irradiation–induced point defects in the CuO_2–planes. Only one model consistently describes all aspects of the reduction of the c-axis superfluid density with temperature and disorder strength. This is the Impurity Assisted Hopping model of Radtke *et al.*,[27] elaborated upon by Xiang and Wheatley,[23,25] and by Kim and Carbotte.[11] The model supposes a reduction of $\rho_s^c(T)$ through hopping of nodal quasi-particles assisted by weak, highly anisotropic scattering by defects in the insulating SrO and BiO layers. Candidates for such impurities are out-of-plane oxygen defects[19] or cation disorder. From a fit of $f_{pl}(T)$ to the IAH model, we extract the energy scale $\Delta_0 \sim 2.5k_BT_c$ characterizing nodal quasiparticle excitations. This is remarkably close to the number obtained by Le Tacon *et al.* from anisotropic Raman scattering,[53] giving further confidence in the IAH interpretation.

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